SFE-IR Analysis of Water in Plutonium-bearing Process Solids

Application Overview

Residual water is a critical concern in spent nuclear fuels and other plutonium-bearing process solids. Due to radiolysis, potentially explosive hydrogen gas can be generated from water.

US Department of Energy (DOE) STD-3013-99 provides criteria for stabilization of plutonium-bearing materials at DOE facilities to safe and stable forms that can be packaged and placed in storage with minimal surveillance for up to 50 years. This Standard applies to plutonium-bearing metals and oxides containing at least 30 wt% plutonium plus uranium.

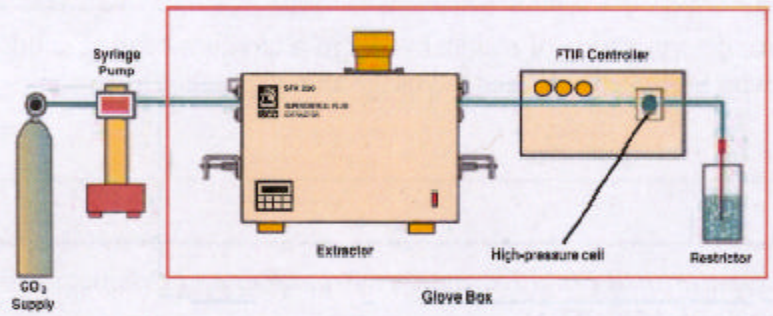
DOE has approved three analytical methods for determining water in these materials:

- Thermogravimetric Analysis
- Neutron Scattering Analysis
- Online SFE-IR.

Thermogravimetric analysis (LOI or "loss on ignition") has been the traditional method for determining water content in spent fuel samples. Unfortunately, volatile salts commonly found in fuels can interfere with this analysis. Neutron scattering analysis is accurate but expensive, since a neutron source is needed. SFE with online FTIR analysis is accurate, specific to water, and affordable.

SFE-IR Method

SFE-IR, as shown below, uses a high-pressure pump to provide a constant flow of dry, supercritical carbon dioxide (CO_2) through samples contained in a heated extraction chamber. The CO_2 acts as a carrier fluid, transporting solvated water to a standard Fourier Transform Infrared (FTIR) detector equipped with special high-pressure cell. The amount of water in the supercritical CO_2 stream is quantified using unique infrared absorbance bands, e.g. near 1600cm^{-1}, which allows SFE-IR to be specific to water. The FTIR response is calibrated against an injection of a known amount of water.



SFE-IR system for monitoring water in plutonium-bearing solids. The compact extractor module and FTIR spectrometer are contained inside the glovebox.

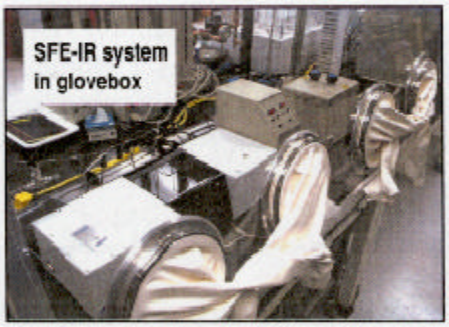
Advantages of the SFE-IR method include:

- The sample size (nominally 5 g, with a maximum of approximately 20 g) is large enough to cope with most heterogeneous materials.
- Average analysis time of 30 - 60 minutes with real-time data acquisition and software-automated.quantitation. Shorter analysis time is possible if a qualitative (< 0.5 wt. % or > 0.5 wt. %) result is acceptable.
- Minimal waste generation (< 1 liter of liquid per year).
- The method and operation have DOE LAAO approval for use in a CAT-1 nuclear facility.

Instrumentation & facilities

Instrumentation and facilties for the SFE-IR analysis consist of:

1. SFE system including high-pressure pump and semi-automated extractor *
2. Heated transfer line and high-pressure SFE-IR cell *
3. Standard FTIR spectrometer with computer and software
4. Standard double glovebox with feed-through ports for CO_2 line and data cable, plus feed-through port or open-front box to transfer materials and samples into the glovebox



* These instruments are available from Isco, Inc. For complete specifications, ordering, and delivery information, please contact Isco at: **(800)228-4250** (toll-free USA and Canada), by Fax at (402) 465-3022, or via e-mail to **info@isco.com**.

Summary

SFE-IR is a DOE approved method for determination of residual water in plutonium-bearing solids. Quantitative results can be obtained with high precision, and the usage and disposal of hazardous solvents are minimized.

References

M. A. McHugh and V.J. Krukonis, *Supercritical Fluid Extraction : Principles and Practice*, 2nd edition (Butterworth Publishers, Stoneham, MA, 1994).